Dynamic ion-ion structure factor of strongly coupled hydrogen plasmas at arbitrary degeneracies

Filippo Nardin and Gianni Jacucci
Department of Physics, University of Trento, I-38050 Povo, Trento, Italy

M. W. C. Dharma-wardana
National Research Council of Canada, Ottawa, Canada K1A 0R6
(Received 1 September 1987)

Results are reported for the dynamic structure factor $S(k\omega)$ for ions in the hitherto theoretically inaccessible regime of strongly coupled partially degenerate hydrogen plasmas where the electron-ion interaction is not weak. Results from density-functional theory and molecular dynamics have been used to provide a rigorous approach which avoids linear-response approximations or the construction of pseudopotentials. The results are discussed in terms of a microscopic theory and a hydrodynamic model. Values of the sound velocity, thermal diffusivity, the specific-heat ratio, and the viscosity for these plasmas are reported.

A hydrogen plasma is one of the simplest condensed-matter systems which is also a real system of great practical interest in many diverse fields such as astrophysics, semiconductor physics, and in inertial fusion. The static structure factor $S(k)$ and the dynamic structure factor $S(k\omega)$ of the ions are of great importance in describing bremsstrahlung, light scattering, screening and other properties of a hydrogen plasma. Nevertheless, systematic theoretical methods are not available except in the limit where all particles can be assumed to behave as classical points, or in the one-component plasma (OCP) limit where the electrons can be modeled by a uniform static background.[1-4] In the OCP the electron-proton system is reduced to an effective one-component classical problem where the interaction $U(r)$ between the ions is simply the Coulomb potential. A slightly more realistic model is obtained by allowing the electrons to respond linearly to the ions.[5] Then we have a classical problem in which $U(r)$ is the linearly screened Coulomb interaction, viz., $V(q)/\epsilon(q)$, where $V(q)$ is the Coulomb potential $4\pi/q^2$, and $\epsilon(q)$ is the electron gas dielectric function. This too can be handled by molecular dynamics simulation,[5] once a form for $\epsilon(q)$ is chosen. In this approach the electron gas correlations are embedded in $\epsilon(q)$, and molecular dynamics (MD) is used to deal with the ion-ion correlations. This approach assumes that the electron-proton interaction is weak, and is useful in a restricted regime of (very high) densities and (low electron) temperatures. A two-component plasma model, where both the electrons and the ions are treated as classical (and with the electron-ion interaction still assumed to be weak), can also be studied by MD if suitable (if somewhat artificial) cutoffs are introduced to prevent classical collapse of electron-ion orbits.[4] Here there are problems as to which pair interaction should be used,[6] and the MD simulation yields data on $S(k\omega)$ for the electrons, but *not for the ions,* due to the differences in time scales for these two species.

Hence, it is clear that a systematic method capable of calculating $S(k\omega)$ for plasmas, where (a) the electrons are of arbitrary degeneracy and (b) the electron-ion interaction is not weak, is needed. At this point we note that, as far as static correlations are concerned, a rigorous first-principles method is available via density-functional theory (DFT). That is, the ion-ion structure factor $S(k)$ for an arbitrary plasma can be calculated using the method given in Ref. 7. There, the ion-density profile $\rho(r)$ around a given ion at the origin, taken as the "external field," is calculated self-consistently, together with the associated neutralizing electron density profile $n(r)$, using a pair of coupled density-functional equations for the ions and electrons, respectively. If the average ion density is $\bar{\rho}$, the ion-ion pair correlation function $g(r)$ is equal to $\rho(r)/\bar{\rho}$. This incorporates the nonlinear screening effects implicit in the self-consistent solution of the Kohn-Sham equations. Once $g(r)$ is known [see Fig. 1(a)], $S(k)$ is available through a Fourier transformation. In Ref. 7 it was shown that this procedure leads to good agreement with known MD results for $g(r)$ in the relevant limits.

At this point we ask if the rigorously obtained information regarding the static correlations in the system contained in $S(k)$ could be used to develop an $S(k\omega)$. This is very much in the spirit of renormalized kinetic theory where dynamic correlations are constructed assuming that the static correlations are known. Our approach is to note that the $g(r)$ for the ions obtained from DFT defines an effective ion-ion interaction via the relation

$$g(r)=\exp[-\beta U(r)+N(r)+B(r)] , \tag{1}$$

where $\beta=(k_B T)^{-1}$ is the inverse temperature in atomic units and $N(r),B(r)$ are the nodal and Bridge diagram contributions.[8] The hypernetted-chain (HNC) approximation is to set $B(r)$ to zero. Then $N(r)$ can be expressed in terms of $g(r)$ and hence $U(r)$ can be determined[8,9] from a knowledge of $g(r)$ only [see Fig. 1(b)]. If we define the strong-coupling plasma parameter Γ_{ii} for the ions as the mean potential energy divided by the

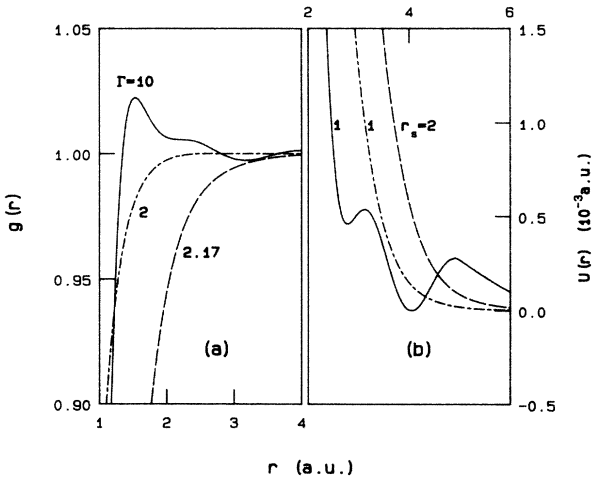

FIG. 1. (a) shows $g(r)$ for $r_s=1$, $\Gamma=10$, solid line; $r_s=1$, $\Gamma=2$, dashed-dotted line; $r_s=2$, $\Gamma=2.17$, dashed line. (b) shows $U(r)$ obtained by inversion of the $g(r)$.

kinetic energy, then $\Gamma_{ii}\approx\beta U(r_0)$, where r_0 is the mean ion-sphere radius, such that $4\pi r_0^3/3=\bar{\rho}$. The HNC approximation is known to be excellent for $\Gamma_{ii}\leq 15$ and hence certainly applicable to the hydrogen plasmas studied here. Another important coupling parameter, Γ_{ie}, is defined by the electron-ion interaction V_{ie}, which is attractive and could generate electron-ion bound states. For weak V_{ie} and small Γ_{ii} the pair-potential $U(r)$ obtained from the inversion of (1) would prove to be nothing but the linearly screened Coulomb potential. But for strong V_{ie} the pair potential $U(r)$ would be a nonlinearly screened Coulomb potential which would have to be determined from (1) for each temperature and density. We should also emphasize that "three-body" effects, etc., are not necessary in the regime of Γ_{ii} studied here. Indeed, even if three-body effects were present they would merely modify the exact two-body term to give an effective two-body potential defined via (1).

Thus the method exploited in this paper is as follows. We carry out a density-functional calculation for the plasma along the lines given in Ref. 7. The resulting ion-ion pair correlation function is inverted[8,9] via Eq. (1) to obtain the nonlinearly screened effective ion-ion pair potential. This $U(r)$, containing nonlinear *static* screening can be consistently used for obtaining $S(k\omega)$ for ions since the ω entering into ion dynamics is much smaller than those for electrons (i.e., $\omega\ll\omega_p^e$ where ω_p^e is the electron plasma frequency). The $S(k\omega)$ is obtained from a MD simulation for the ions interacting via the effective pair potential $U(r)$. That is, the two-component problem has been replaced by an effective one-component problem. Note that this procedure is similar in spirit to that used in the theory of liquid metals[10] to reduce the two-component problem to an effective one-component form. However, the usual theory[10] invokes linear response (LR) and hence the methods used here can be exploited for liquid metals (e.g., transition metal liquids) for which LR is inadmissable.

Having obtained $S(k\omega)$ from MD using the $U(r)$ deduced from the $g(r)$ of DFT, we are now in a position to look for microscopic and other models for $S(k\omega)$ for this system. Here we consider hydrogen plasma with electron densities n and ion densities $\rho=n/Z$, $Z=1$, such that the electron-sphere radius $r_s=1$ and 2 a.u. We also consider temperatures $T=\beta^{-1}$ such that the strong-coupling plasma parameter $\Gamma=\beta/r_s$ ranges from 2–10. The lower density ($r_s=2$) high-temperature (small-Γ) regime is very *close to the onset of bound states.* In the examples presented here, the degeneracy parameter $\tilde{T}=T/T_F$, where T_F is the Fermi temperature of the electrons, ranges from 0.05–0.5.

We report results for $S(k\omega)$ from MD using the pair interactions obtained from DFT, from a simple microscopic theory with no adjustable parameters, and from the macroscopic hydrodynamic form of $S(k\omega)$ where the parameters are fitted to the MD data. The latter model enables us to directly extract results for the ratio of the specific heats γ, thermal diffusivity D_T, kinematic viscosity b, sound velocity c_s, and the sound attenuation coefficient γ_s for this hitherto inaccessible regime of plasmas.

The microscopic model, to be called the local-field model (LFM), is the well-known generalized linear-response form first suggested by Hubbard.[11] The ion-response function $\chi(k\omega)$ is related to the dynamic structure factor by

$$S(k\omega)=-\mathrm{Im}\chi(k\omega)/\pi\beta\omega\ . \tag{2}$$

The response function can be expressed as[11]

$$\chi(k\omega)=\chi^0(k\omega)/[1-Z^2V_k(1-G_k)\chi^0(k\omega)]\ . \tag{3}$$

Here Z is the ionic charge, $V_k=4\pi/k^2$ is the Coulomb potential, and $\chi^0(k\omega)$ is the free-particle response function for the classical ions, such that

$$\mathrm{Im}\chi^0(k\omega)=\frac{-k_D^2}{4\bar{k}\sqrt{\pi}}\exp\left[-\frac{\bar{\omega}^2}{4\bar{k}^2}+\frac{\bar{k}^2}{4}\right]\sinh(\bar{\omega}/2)\ . \tag{4}$$

In Eq. (4) $\bar{k}=k/k_{\mathrm{th}}$, where the thermal momentum k_{th} is given by $k_{\mathrm{th}}^2=2M/\beta$, $\bar{\omega}=\omega\beta$, M is the ion mass, and k_D is the Debye momentum such that $k_D^2=4\pi Z^2\rho\beta$, with ρ the ion density. Equation (4) is sufficient to define the full $\chi^0(k\omega)$ since the real part is given from (4) by a Kramers-Kronig relation. Note that the dynamic correlations in the local field are neglected and a static form, viz., G_k, is used in (3).

The static correlations in the proton-electron two-component system have been rigorously treated in the LDA-DFT calculation. Hence, the static local field G_k occurring in (3) is chosen to reproduce the static structure factor $S(k)$ obtained from the DFT calculation. Thus

$$\int_{-\infty}^{+\infty}S(k\omega)d\omega=\rho S(k)_{\mathrm{DFT}}\ , \tag{5}$$

$$G_k=1+\frac{k^2}{k_D^2}[1-1/S(k)_{\mathrm{DFT}}]\ . \tag{6}$$

Equation (3) uses a *static* local field G_k. The extent of the validity of such models for these plasmas is unknown. Indeed *one of the objectives of this study* is to

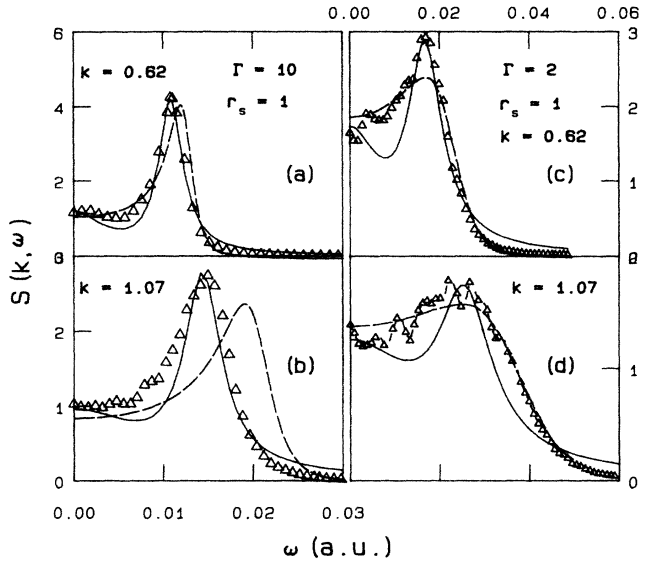

FIG. 2. (a) and (b) are for $r_s=1$, $\Gamma=10$ at $k=0.62$ and 1.07 a.u., respectively; (c) and (d) are for the $\Gamma=2$ case. The triangles are from MD. The solid line is the hydrodynamic fit, while the dashed line gives the local-field model.

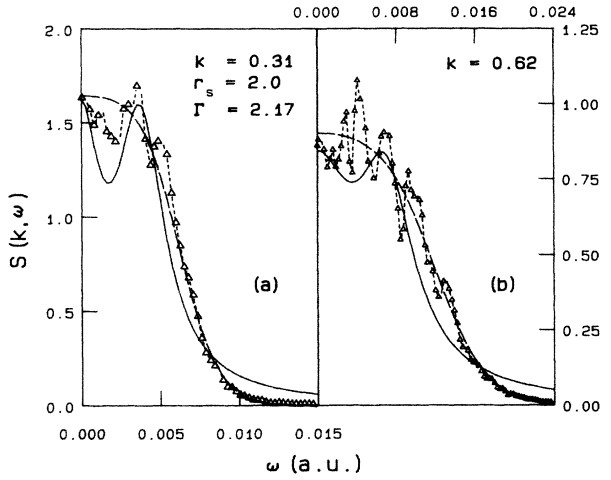

FIG. 3. As in Fig. 2 but for $r_s=2$, $\Gamma=2.17$ and $k=0.31$ and 0.62 a.u. in (a) and (b), respectively.

test them against the MD results for $S(k\omega)$.

A hydrogen plasma is a charge-compensated real fluid (rather than an OCP) where the ion-plasma frequency gets (screened and) converted to an acoustic mode. Hence, much of the physics should be recoverable by a hydrodynamic model[12,13] of $S(k\omega)$ for $\omega<\omega_p^e$ and for small k. In the hydrodynamic model (HDM) of $S(k\omega)$ we write

$$S(k\omega)=\frac{\rho S(k)}{2\pi}\left\{\frac{2(\gamma-1)}{\gamma}\frac{\Delta R}{\omega^2+(\Delta R)^2}+\frac{1}{\gamma}\frac{\Delta B}{(\omega+\omega_B)^2+\Delta B^2}+\frac{1}{\gamma}\frac{\Delta B}{(\omega-\omega_B)^2+\Delta B^2}\right\}, \tag{7}$$

where ΔR and ΔB are the half widths of the Rayleigh and Brillouin peaks, respectively. The Brillouin frequency ω_B is simply the screened ion-plasma frequency (sound mode). In the hydrodynamic regime we have the dispersion relations of the form

$$\omega_B=c_s k,\quad \Delta R=D_T k^2,\quad \Delta B=\gamma_s k^2, \tag{8}$$

where the thermal diffusivity D_T is related to the sound attenuation coefficient γ_s and the viscosity b by the relation $\gamma_s=0.5(\gamma D_T^2 k^2+b)$. The model may be expected to be valid in a more general sense[12] if the simple dispersion relations are not invoked. Then we have four adjustable parameters γ, ΔR,ω_B, and b with the constraints $\gamma>1$, $b>0$.

The results for $S(k\omega)$ for $r_s=1$, $\Gamma=10$, and $k=0.62$ and 1.07 a.u. are shown in Figs. 2(a) and 2(b), respectively. The agreement between MD (triangles) and LFM (dashed line) for $k=0.62$ is surprisingly good considering that LFM is a theory without adjustable parameters. This agreement is destroyed at $k=1.07$. Figures 2(c) and 2(d) show the results for $\Gamma=2$, and $k=0.62$ and 1.07 a.u., respectively. Here it is rather surprising to note that the collective mode is *more damped* in the LFM where there is no contribution to the damping from the (static) local field. The rapid decay of $S(k\omega)$ for large ω is well described by LFM and, as expected, rather poorly by the ω^{-2} dependence found in HDM (solid curve).

Figures 3(a) and 3(b) present results for the lower density lower degeneracy case $r_s=2$, $\Gamma=2.17$ ($\tilde{T}=0.5$) for $k=0.31$ and 0.62 a.u., respectively. The MD data (triangles) show much more structure than the theoretical model calculations. The LFM provides a good average representation to the data. The physical constants obtained from the hydrodynamic fits at $k=0.62$ a.u. for the three plasmas are given in Table I, assuming the simple dispersion relations given by Eq. (8). A more reliable extraction of the physical constants and dispersion relations will require more elaborate modeling or the construction of microscopic theories superior to the HDM and LFM discussed here. A more detailed discussion

TABLE I. The isentropic exponent γ, thermal diffusivity D_T, and the sound velocity c_s as obtained from the hydrodynamic model fits at the lowest momentum (k) studied. The viscosity $b\simeq 10^{-6}$ in all cases. Atomic units are used. Thus $c_s=0.0174$ a.u. is 1.46×10^6 cm/sec.

r_s	Γ	k	γ	$10D_T$	$10c_s$
1	10.0	0.62	1.31	0.103	0.174
1	2.0	0.62	1.45	0.202	0.271
2	2.17	0.31	1.27	0.136	0.121

will be taken up elsewhere.

In conclusion, we have shown how essentially any arbitrary fluid with electrons and nuclei can be described by an effective ion-ion interaction defining an effective classical fluid for $\omega < \omega_p^e$, taking electron degeneracy effects, strong electron-ion, electron-electron interactions into account with only the LDA as the essential approximation in a DFT calculation. In this approach it is not necessary to explicitly construct pseudopotentials or screening. The static and dynamic properties of such systems can then be calculated by straightforward methods valid for simple fluids, as illustrated by our results for $S(k\omega)$. Results for $S(k\omega)$ are immediately useful for understanding plasma bremsstrahlung,[14] transport coefficients, lifetime effects, etc. Also, the methods presented here will have implications in the theory of ion-ion interactions[15] in liquid transition metals and in the study of fast processes in electron-hole plasmas in semiconductors.[16]

We acknowledge valuable discussions with Marco Ronchetti (Trento) and Bob Cauble (Lawrence Livermore Laboratory). The Department of Physics at the University of Trento is part of the Centro Studi del Consiglio Nationale delle Ricerche.

[1]S. Ichimaru, S. Mitake, S. Tanaka, and X-Z. Yan, Phys. Rev. A **32**, 1768 (1985).

[2]R. Cauble and D. B. Boercker, Phys. Rev. A **28**, 944 (1983).

[3]See J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic, New York, 1976), Chaps. 7 and 8.

[4]J.-P. Hansen and I. R. McDonald, Phys. Rev. A **23**, 2041 (1981).

[5]H. E. DeWitt and W. B. Hubbard, Astrophys. J. **205**, 295 (1976), S. Galam and J. P. Hansen, Phys. Rev. A **14**, 816 (1976); S. Tanaka and S. Ichimari, J. Phys. Soc. Jpn. **53**, 2039 (1984).

[6]J. Wallenborn, V. Zehnlé, and B. Bernu, Europhys. Lett. **3**, 661 (1987).

[7]M. W. C. Dharma-wardana and F. Perrot, Phys. Rev. A **26**, 2096 (1982). For a review see *Strongly Coupled Plasma Physics,* edited by H. E. DeWitt and F. Rogers (Plenum, New York, 1987), pp. 275–292.

[8]M. W. C. Dharma-wardana, F. Perrot, and G. C. Aers, Phys. Rev. A **28**, 344 (1983).

[9]M. W. C. Dharma-wardana and G. C. Aers, Phys. Rev. Lett. **56**, 1211 (1986).

[10]N. W. Ashcroft and D. Stroud, Solid State Phys. **33**, 1 (1978).

[11]J. Hubbard, Proc. R. Soc. London, Ser. A **243**, 336 (1957).

[12]M. Baus and J.-P. Hansen, Phys. Rep. **59**, 43 (1980).

[13]J. P. Boon and S. Yip, *Molecular Hydrodynamics* (McGraw-Hill, New York, 1980).

[14]L. Kim, R. H. Pratt, and H. K. Tseng, Phys. Rev. A **32**, 1693 (1985).

[15]A. E. Carlsson, Phys. Rev. B **32**, 4866 (1985).

[16]R. A. Höpfel, J. Shah, and A. Gossard, Phys. Rev. Lett. **56**, 765 (1986).